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# Growth of GaAs Thin Film Via Supersonic Beams of a Single Source Molecular Precursor

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## ABSTRACT

The growth of GaAs thin films was achieved using beams of a single source molecular precursor (  $(t\text{-Bu})_2(\text{Ga-As})(t\text{-Bu})_2$  ). In this study supersonic beams of this molecules were produced after seeding in Helium. We were able to produce GaAs thin films at rates of  $0.05\text{ }\mu\text{m/h}$  with a nozzle aperture of  $50\text{ }\mu\text{m}$  and a helium stagnation pressure of 200 torr. These growth rates were dramatically higher than those produced from effusive beams of the single source molecular precursor. The growth experiments on substrates held at 500, 450, 400 and  $300\text{ }^\circ\text{C}$  produced good crystalline materials and relatively smooth film surface as inferred from RHEED and AFM

## I Introduction

In this letter we describe the first growth experiment of GaAs thin films using supersonic molecular beam of a single source molecular precursor. Thin film growth of GaAs was achieved by directing beams of  $(t\text{-Bu})_2(\text{Ga-As})(t\text{-Bu})_2$  [1] seeded in Helium at  $60^\circ\text{C}$  incidence angle onto GaAs (100) substrates heated to various temperatures.

A supersonic beam [2] is simply produced by allowing a gas to expand very rapidly from a high pressure (50 - 200 torr) chamber (nozzle) through small aperture (50-200  $\mu\text{m}$ ) into a low pressure chamber (the growth chamber in our study). The mean free path of the molecules inside the nozzle is much smaller than the diameter of the nozzle orifice for supersonic beams, in contrast to the effusive beams used in molecular beam epitaxy (MBE) and Chemical Beam Epitaxy (CBE), for which the atomic or molecular mean free path is usually much greater than the aperture diameter. For this reason the supersonic expansion offers beams with very special properties that are not achievable with conventional effusive beams.

The most important properties of the supersonic free expansion for film growth reside in (1) the highly monochromatized and collimated beam which are the consequence of the conversion of the random motion in the nozzle into a highly directed motion perpendicular to the nozzle aperture during the free expansion of the molecular in the low pressure region, (2) the enhanced beam intensities as a consequences of collimation and monochromatization of the beam. These intensities can be a couple of collimation orders of magnitude higher than the intensities of effusive beams, (3) The tuning of the kinetic energy of the supersonic molecular beams that can be achieved by controlling the gas temperature in the nozzle or using seeding techniques. In this techniques small amounts of heavy molecules are transported in a light carrier gas and are brought to superthermal velocities in the free expansion of the gas mixture [2]. (4) The potential enhancement in the growth rates through more efficient dissociation of molecules accelerated to superthermal velocities [3, 4]. With a molecular mass of 373

molecules the average kinetic energy of the single source precursor  $((t\text{-Bu})_2(\text{Ga-As})(t\text{-Bu})_2)$  molecules is approximately 6.0 eV

The single source precursor  $(t\text{-Bu})_2(\text{Ga-As})(t\text{-Bu})_2$  used in this study was synthesized By Higa and George<sup>[1]</sup>. At room temperature this compound is solid with a vapor pressure of  $10^{-1}$  torr. Its sublimation temperature is 45 °C. Pyrolysis studies of this compound show that decomposition begins to take place at 158 °C. This compound is already being used in CBE experiments by Ekerdt 's group <sup>[5, 8]</sup> at the Chemical Engineering Department of the University of Texas at Austin. Their results show growth rates of  $\approx 0.1 \mu\text{m}/\text{hour}$  and good crystalline GaAs thin film above 500 °C growth temperatures .

## II Experiment

Our growth studies were conducted in a small scale MBE system <sup>[9]</sup> composed of a growth chamber and a load lock chamber. The growth chamber is equipped with a mass spectrometer, a RHEED system and commercial Ga and As sources. The base pressure in the growth chamber was  $1 \times 10^{-9}$  torr. The deposition experiments were conducted on a semi-insulating GaAs wafer with an area of  $3\text{-}4 \text{ cm}^2$ . The GaAs (100) surface was cleaned by annealing to 560 -600 ° C in an As pressure of  $10^{-6}$  torr for a few minutes.

The supersonic beam doser consists of a 2" stainless nipple (for loading the single source precursor ) connected to a 2 3/4 CF flange by a metal valve on one side and to a gas manifold on the other side. The 2 3/4 CF flange supports a nozzle made of 1/4 OD stainless steel tube which extends about 10 " into the growth chamber. In this arrangement the axis of the nozzle was at 60° off the normal to the GaAs (100) surface. The aperture of the nozzle was 50  $\mu\text{m}$ . In addition, the 2 3/4 flange is placed on a linear motion manipulator to allow the tip of the nozzle to be adjusted at a desired distance from the GaAs wafer. The nozzle aperture was placed at  $\approx 2 \text{ cm}$  from the substrate . In

this setting the nozzle and the doser parts outside the vacuum can be heated uniformly to a desired temperature.

In all our growth experiments the He pressure in the nozzle (and the doser) was maintained at 150 torr. The doser and the nozzle were both kept at a temperatures near 80° C. The pressure in the growth chamber was near  $2.0 \times 10^{-4}$  torr. A mesh made of .010" tantalum wire covered most of the GaAs wafer. This generates steps between growth areas and areas covered by the tantalum wires (part of the mesh). By measuring the step heights with a profilometer, GaAs thin film thicknesses and growth rates can be estimated.

### III Results and Discussion

The most important result in this study of the growth GaAs thin films with supersonic beams of single source precursor molecules  $(t\text{-Bu})_2(\text{Ga-As})(t\text{-Bu})_2$  is the dramatic enhancement in growth rates relative to those obtained in effusive beam growth experiment. This is the experiment in which the single source precursors is delivered through the nozzle aperture with no carrier gas in the nozzle. In fact, after dosing for 8 hours the surface of the GaAs substrate showed no measurable film thickness with the profilometer. In contrast single source precursor molecular beams delivered after seeding in Helium produced GaAs thin film growth at a rate of 0.05  $\mu\text{m}/\text{hour}$  near the center of the sample which was defined by the intersection of the axis of the nozzle and the plane containing the GaAs(100) surface. The growth rate dropped to zero within 1 to 1.5 cm from the center of the sample.

Thin films of GaAs deposited at 500, 450, and 400 °C substrate temperatures show a sharp RHEED pattern which was of the same quality as that shown by the GaAs(100) surface prior to deposition. In contrast, the GaAs thin film deposited at 300 °C substrate temperature yields a RHEED pattern which was more diffuse. The RHEED observations on thin films deposited on GaAs substrates held at 500, 450 and 400 °C

indicate the GaAs growth is epitaxial. However, the growth at 300 °C substrate temperature appears to yield films of poorer crystallinity .

The morphology of the GaAs thin film was examined with AFM (Atomic Force Microscopy)[10]. At all deposition temperatures the surface quality of the films was better than that of the GaAs substrate after removal of the surface oxide by annealing to 560 -600 °C in an As overpressure. The rms (root mean square) of the vertical displacement of the AFM tip is one parameter which can be used to quantify surface roughness. We obtained an rms value of 8.1 Å for the GaAs thin film deposited at 450 °C and an rms value of 11 Å for the GaAs substrate which was annealed in an As overpressure. For a GaAs substrate which was not treated in vacuum this value was 2.4 Å which corresponds to an extremely smooth surface. This simply indicates that our sample preparation in vacuum prior to film deposition induces more surface roughness on the GaAs (100) surface. The deposition of the GaAs thin film, however, appears to reduce roughness. Further studies are required using smoother GaAs surface prior to film deposition in order to establish any correlation between surface quality in terms of roughness and growth conditions (substrate temperature, beam intensities ).

In conclusion we have shown in this letter that dramatically faster growth rates can be achieved using single source molecular beams of  $(t\text{-Bu})_2(\text{Ga-As})(t\text{-Bu})_2$  seeded in 150 torr of Helium in comparison to effusive beam experiments. In addition a lower limit near 300 °C for the epitaxial growth temperature of GaAs thin from this single source precursor was determined.

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